Proceedings of the American Academy of Arts and Sciences.

Vol. 63. No. 7.—December, 1928.

THE EFFECT OF PRESSURE ON THE RESISTANCE OF THREE SERIES OF ALLOYS.

By P. W. Bridgman.

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Presented Oct. 10, 1928. Received Oct. 22, 1928.

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Introduction.

There are many phenomena connected with the electrical behavior of alloys which are difficult to understand. For several years one of the chief reasons for my dissatisfaction with the theory of metallic conduction on which I have been working has been the anomalous behavior of a number of alloys; the behavior of pure metals is more satisfactorily accounted for. It is therefore important to extend our knowledge of the electrical properties of alloys as much as possible; the measurements on the three series of alloys presented here are a contribution in this direction.

Materials and Experimental Methods.

For the series of Fe-Co alloys I am much indebted to Mr. W. C. Ellis,[1] who prepared the alloys and investigated many of their electrical properties as part of a thesis done at Rensselaer Polytechnic Institute under the direction of Dr. M. A. Hunter. The value of Ellis's work is much increased by an X-ray determination which he made of the crystal structure of all his alloys. The series consisted of eight different alloys. Six of these were in the form of drawn wires. Two of them, which could not be drawn, were in the form of swaged rods about 6 mm. in diameter; slender pieces suitable for the measurements under pressure were prepared from these rods by grinding. A special feature of the material prepared by Ellis is the high purity of his cobalt, which was electrolytic. Its high purity is evidenced by the low specific resistance and high temperature coefficient, as given by Ellis in his paper. A remarkable feature of the series is the fact, emphasized by Ellis, that the

specific resistance of the alloys from 32 to 53 atomic per cent Fe have a lower specific resistance than either of the pure constituents.* So far as I know, this is the first example of such behavior in alloys; in fact one can frequently find the generalization made in the literature that the resistance of an alloy is greater than that of either component. As Ellis remarks, this effect may be expected to be of significance in theories of metallic conduction. Certainly such behavior lends great interest to as complete a knowledge as possible of the properties of this series of alloys.

The series of Fe-Ni alloys I owe to the kindness of Dr. L. W. McKeehan, at that time with the Bell Telephone Laboratories, Inc. He had a series of 15 alloys prepared and drawn to wire. The following description of the alloys is quoted from Dr. McKeehan's letter. "These wires are all hard drawn. In order to check the approximate composition and to get some idea of the variation of resistivity with composition, I have had the resistivity measured in the state furnished, and also on short samples that have been annealed at 1000° in vacuum for one hour and allowed to cool slowly, *i. e.*, requiring several hours to cool to room temperature. The results are given on the attached photostat (reproduced in Figure 1). In regard to the exact composition, I have not figures for all the alloys, but on those that have been analyzed the actual percentage of iron or nickel falls within less than ½% of the nominal composition."

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* I am not sure that this result can be regarded as established beyond all possibility of doubt. It is evident that the resistance of Co is extraordinarily sensitive to slight impurities; thus Kalmus[3] found for the specific resistance of pure Co reduced in hydrogen the value 8.96×10^{-6} at 20°, against 6.24 found by Ellis. The value of Ellis might possibly be materially reduced if there were a very slight amount of remaining impurity. The reduction need not be great in order to change Ellis's conclusion, because he finds for the resistance at 0° of his pure Co 5.60×10^{-6}, whereas the two alloys with lowest resistance, containing 33.3% and 50% Fe respectively, had resistances of 5.15 and 5.13. It is significant in this connection that Grüneisen[3] has indicated as the most probable value for the resistance of pure Co, on the basis of computations from the resistance at very low temperatures, made by means of Matthiesen's rule, on samples known to be impure, the figure 5×10^{-6}. Furthermore, it would appear that purer cobalt than that of Ellis has been prepared, for Holborn[4] gives for the temperature coefficient between 0° and 100° of pure Co 0.00658, against Ellis's 0.00604. Holborn does not give the specific resistance of his material.

In addition to the more or less complete results on these two series of alloys, I am taking this opportunity to publish some partial results obtained with my high pressure apparatus by Mr. J. R. Oppenheimer in 1925 on the Cu-Ni series. These results cover a range of composition up to about 50% Ni. The material was electrolytic copper from the Bureau of Standards, and commercially pure nickel

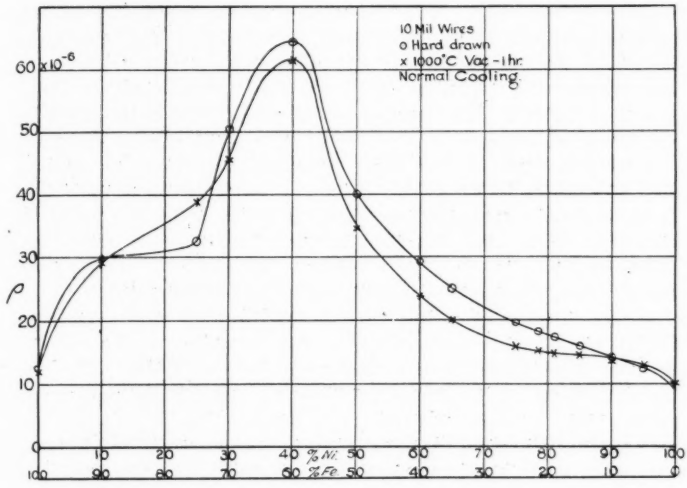

FIGURE 1. The specific resistance in the Fe-Ni series as a function of composition in the hard drawn condition, and after annealing.

of probably about 99% purity. The alloys were prepared by melting together in a vacuum in a quartz tube weighed amounts of the two constituents. Floating on the upper surface of the melted metal was a quartz capillary perhaps 8 cm. long and 0.5 mm. in diameter, closed at the upper end. When the melting was completed and the metals thoroughly mixed, air or nitrogen was suddenly admitted to the quartz tube, driving the molten metal up into the capillary, and thus producing a fine cast wire of the alloy. The wire so obtained was not subjected to any systematic heat treatment, but was used as it came from the tube, after cracking away

the quartz. In general the capillary was always at a temperature lower than the melting point, so that the wire must have experienced a very rapid chilling from the melting temperature to several hundred degrees below.

The pressure measurements were made by the same method as that used in former work,[5] and no detailed description is necessary. The potentiometer method adapted to the measurement of small resistances was used. The description in the remainder of this paragraph applies to the Fe-Co and Fe-Ni series. The wire specimens were bent to the form of a hairpin, about 8 cm on either arm, and current and potential terminals were soft-soldered to either arm near the end. The changes of resistance listed in the following are measured changes; to convert to changes of specific resistance a correction equal to the linear compressibility must be applied. At a pressure of 10,000 kg. the change of resistance due to the change of dimensions is approximately 0.2%, or of the order of 1/10 of the measured change for the pure metals. The pressure range was 12000 kg./cm.^{2}; readings were made at 30° and 75°. At the lower temperature, pressure was transmitted with petroleum-ether to avoid distortion arising from the viscosity of kerosene under pressure. Before the pressure measurements, two seasoning applications of pressure were made, once to 2000 and once to 12000. The permanent changes of resistance produced by these preliminary applications were in almost all cases very small. This is evidence that no important change of internal composition was produced by pressure, such as an unmixing, and in fact inspection of the phase diagrams of these systems shows that no such effect is to be anticipated. In addition to the pressure coefficient of resistance, the mean temperature coefficient between 0° and 100° was obtained by linear extrapolation of the resistances at atmospheric pressure at 30° and 75°. Since the range 30°–75° is nearly in the middle of the range 0°–100°, linear extrapolation gives approximately the correct mean coefficient, although such an extrapolation would not in general be reliable because the relation between resistance and temperature is not linear. The dimensions of the specimens were also measured, and the specific resistance found. The chief error here is in the diameter of the wire, and should not be more than 1% or 2%. The diameter was measured with an ordinary micrometer, and varied from 0.025 cm. to 0.075 cm.

The measurements of the Cu-Ni series were less elaborate, and

consisted of measurements by the potentiometer method of resistance as a function of pressure up to 12000 kg at 25°.

Experimental Results.

The numerical results of the measurements are given in the following tables. In all cases except one, the relation between pressure and resistance could be reproduced by a second degree formula in the pressure; the two constants of this formula are given in the tables. The one exception is the alloy 70% Fe, 30% Ni; for this the detailed dependence of resistance on pressure is shown in Table IV and in Table II only the mean coefficient to 12000 is given. The tables also contain the specific resistance at 30°, the mean temperature coefficient between 0° and 100°, and the mean deviation of a single pressure reading from a smooth curve in terms of the maximum pressure effect. This last gives an idea of the accuracy of the pressure measurements; of course the percentage deviation is much larger for those alloys with a small pressure coefficient.

Ellis's pure Co is materially purer than most of the Co whose electrical properties are listed in the literature, and much purer than the two samples for which I have previously measured the pressure coefficient.[6] The values given in the table should, therefore, supercede my previous values. Although the temperature coefficient of the new sample is much higher than that of my best previous sample, 0.0060 against 0.0044, the pressure coefficient is not materially different, the average coefficient between 0 and 12000 kg. of the new material being -0.950×10^{-6} against -0.934×10^{-6} found before. The difference is in the direction usually found to go with increasing purity.

The pressure effect on the resistance of Ellis's pure Fe was not measured, since I had previously measured[7] the effect on Fe of approximately the same grade of purity as that of Ellis. Ellis gives the impurity as 0.035%, and my American Ingot iron was stated to have about 0.03% impurity. Ellis's temperature coefficient is somewhat higher than mine, 0.00635 against 0.00621, so that on this ground Ellis's iron was probably somewhat purer than mine, but the difference is probably not sufficient to introduce an appreciable difference in the pressure coefficient. Neither the temperature coefficient of Ellis or myself is as high as the highest listed for iron, namely 0.00657 by Holborn.[8]

The values given in the table for the Fe-Ni series for pure Fe

TABLE I.

RESULTS FOR FE-CO SERIES.

Composition At. % Fe	Co	Effect of Pressure on Resistance $\frac{\Delta R}{R_0} = ap + bp^2$ 30° a $\times 10^6$	30° b $\times 10^{11}$	30° Deviation %	75° a $\times 10^6$	75° b $\times 10^{11}$	75° Deviation %	Specific* Resistance at 30° $\times 10^6$	Mean* Temperature Coefficient 0–100°
0	100	− .958	+ 0.68	1.1	− .960	+1.04	2.7	6.62	.00604
10	90	− .401	+ 0.69	1.7	− .305	+ .81	2.2	8.29	534
20	80	+4.32	+14.	.5	+6.29	+8.0	.6	9.91	563
33.3	66.7	− .796	+ 0.6	.7	− .643	0	.7	5.84	446
50	50	− .473	+ 2.65	10.2	− .467	+1.63	4.0	5.78	425
66.7	33.3	+1.153	+ 3.10	.1	+ .898	+4.61	1.4	10.37	241
80	20	+1.408	+ .73	.3	+1.346	+ .39	.6	19.34	218
90.9	9.1	−1.413	+ .62	.3	−1.360	+ .49	.5	18.12	275
100.	0	−2.427	+ 1.14		−2.450	+1.00		10.52	635

* Values of Ellis.

TABLE II.

Results for Fe-Ni Series.

Composition Wt. % Fe	Ni	Effect of Pressure on Resistance $\frac{\Delta R}{R_0} = ap + bp^2$ 30° a $\times 10^6$	30° b $\times 10^{11}$	30° Deviation %	75° a $\times 10^6$	75° b $\times 10^{11}$	75° Deviation %	Specific Resistance at 30° $\times 10^6$	Mean Temperature Coefficient 0–100°
100	0	−2.427	+1.14		−2.450	+1.00		10.5	.00621
90	10	+0.036	−8.0	5.1	−0.091	−3.00	2.9	30.1	231
75	25	+2.822	− .92	.2	+2.315	− .92	1.9	32.4	262
70	30	+8.7	(See Table IV)	.1	7.7	(See Table IV)	.1	51.4	589
60	40	+9.100	−6.06	.1	+6.910	−7.08	.0	65.6	287
50	50	+5.904	+5.62	.2	+5.422	+3.68	.3	41.9	443
40	60	+3.002	+2.29	.9	+2.886	+1.70	....	30.7	474
35	65	+1.930	+1.09	.2	+1.776	+1.39	.4	25.7	473
25	75	+0.456		2.0	+0.394		3.1	18.9	628
21.5	78.5	−0.129		6.2	−0.181		12.2	18.2	462
19	81	−0.389		3.3	−0.461		4.2	17.1	446
0	100	−1.905	+0.50		−1.925	+0.56		(7.0)	634

TABLE III.

RESULTS FOR CU-NI SERIES.

Composition Wt. % Ni	Cu	Effect of Pressure on Resistance at 25° $\frac{\Delta R}{R_0} = ap + bp^2$ a	b
0	100	-2.03×10^{-6}	$+0.96 \times 10^{-11}$
0.145	99.855	−1.80	+0.71
1.50	98.50	−1.19	+0.38
4.58	95.42	−0.94	+0.21
8.23	91.77	−0.745	+0.10
12.9	87.1	−0.725	+0.10
24.3	75.7	−0.700	+0.12
50.8	49.2	−0.685	+0.10

TABLE IV.

RESISTANCE OF ALLOY 70% FE–30% NI AS FUNCTION OF PRESSURE.

Pressure kg/cm²	30° $\frac{\Delta R}{R(0.30°)}$	75° $\frac{\Delta R}{R(0.75°)}$
3000	.02478	.02114
6000	.05078	.04279
9000	.07751	.06473
12000	.10477	.08696

and pure Ni were taken from previous work.[7,9] It is probable that the end members of this series as supplied by Professor McKeehan had appreciable amounts of impurity. I measured the pressure and temperature coefficients of the nominally pure iron of this series, and found values considerably lower numerically than my best previous values for pure Fe: 0.00485 against 0.00621 for the temperature coefficient, and -2.09×10^{-6} against -2.43×10^{-6} for the pressure coefficient. It therefore seemed safer to use my previous values for both pure Fe and Ni. Slight impurities in the constituents produce by far the largest effects at the end of the series; there is no reason to expect any important error from impurities

in the intermediate members of the series, between 10% and 90%, as given in the table.

Discussion of Results.

It is important to have before us the crystal structure of the various alloys. The data for the Fe-Ni and the Cu-Ni series will be found in International Critical Tables, Vol. I, p. 350–351, and for the series Fe-Co in the paper already quoted of Ellis.

In the Fe-Ni series, the two end terms have different structures, pure Fe being body centered cubic, and pure Ni face centered cubic (both at room temperature). The body centered structure of pure Fe persists up to between 25 and 30 At % Ni. In this range the effect of the addition of Ni is to increase slightly the side of the fundamental cube, that is to decrease the density slightly. The experimental error here is great; roughly the effect is linear with the amount of Ni, 30 At % Ni increasing the grating space from 2.84 to 2.88 Å, or 1.4%. Beyond 25 or 30% Ni the structure becomes face centered cubic, the same as that of pure Ni. The grating space decreases with increasing amount of Ni, roughly linearly from 3.61 at 25% to 3.53 Å at 100% Ni. In each case the effect of the addition of a slight amount of foreign metal is to distend the crystal structure, which is the opposite of the change produced by hydrostatic pressure. In the case of many dilute solutions of ordinary substances the effect of the addition of a small amount of solute is to produce the same change in physical properties as an extra hydrostatic pressure, that is, the effect of the dissolved substance may be likened to that of an increase of internal pressure. In the case of these Fe-Ni alloys, however, the effect is the exact opposite.

In the Fe-Co series the structure is body centered cubic from pure Fe up to about 70 A% Co. The ultimate effect of the addition of Co is to reduce the grating space, the value of which is 2.850 Å for pure Fe and 2.827 for 66.7% Co. The change of dimensions is not linear with composition, however, but up to about 33.3% Co the dimensions are independent of composition within an error of $\pm$ 0.001. The alloys with 80 and 90 A% Co are face centered cubic, with crystal parameters of 3.550 Å and 3.540 respectively. Finally, pure Co at room temperature is hexagonal close packed. As in the case of the Fe-Ni series, the effect of the addition of a small amount of foreign metal to pure Fe is not to produce the same change of dimensions as an external pressure.

The Cu-Ni alloys form an unbroken series of solid solutions, all of face centered cubic structure, the crystal unit varying from 3.605 for pure Cu to 3.527 Å for pure Ni.

The series of alloys measured here have in common the feature that at all compositions the crystal structure is that of a mixed crystal in which different kinds of atoms occur together, more or less haphazard, in the same crystal lattice. This is opposed to the structure of many binary alloys, in which over a range of composition there is a mechanical mixture in various proportions of mixed crystals of different fixed compositions, or of the pure components. In the range of mechanical mixture, the physical properties of such alloys are linear functions of the composition, the relation being that of the ordinary rule of mixtures. The alloys investigated by Ufford[10] had this structure throughout certain ranges, and within these ranges the pressure coefficient and other physical properties were linear functions of composition. Opposed to this, there is no range in which the pressure coefficient of these three series of alloys is a linear function.

The contents of the several tables are reproduced in Figures 2, 3, and 4 in order that the relations may be more easily grasped. The first and perhaps the most important result brought out by the figures is support of a generalization already made by Ufford,[10] namely that in all known cases the effect of adding a small amount of foreign metal to a pure metal is to increase algebraically its pressure coefficient of resistance. This is shown in the figures by the rise of the curve for pressure coefficient at either end on leaving the pure metal. This generalization is without known exception, and applies not only to these three alloys, but also to the three of very different type investigated by Ufford, and also to several series investigated by Lisell[11] and Beckman.[12] These new results are sufficient to show, however, that the explanation given by Ufford of this general effect cannot always be correct. Ufford pointed out that in the case of his alloys the addition of a small amount of foreign metal compresses the lattice; this is also the effect of hydrostatic pressure. This means, since in practically all cases hydrostatic pressure increases the pressure coefficient algebraically, that the effect of adding a foreign metal is equivalent, in the cases investigated by him, to an increase of external pressure. That this is not true in general is shown by the Fe-Ni series. Here the addition either of Ni to pure Fe or of Fe to pure Ni distends the structure, thus producing the same change of dimensions as that brought

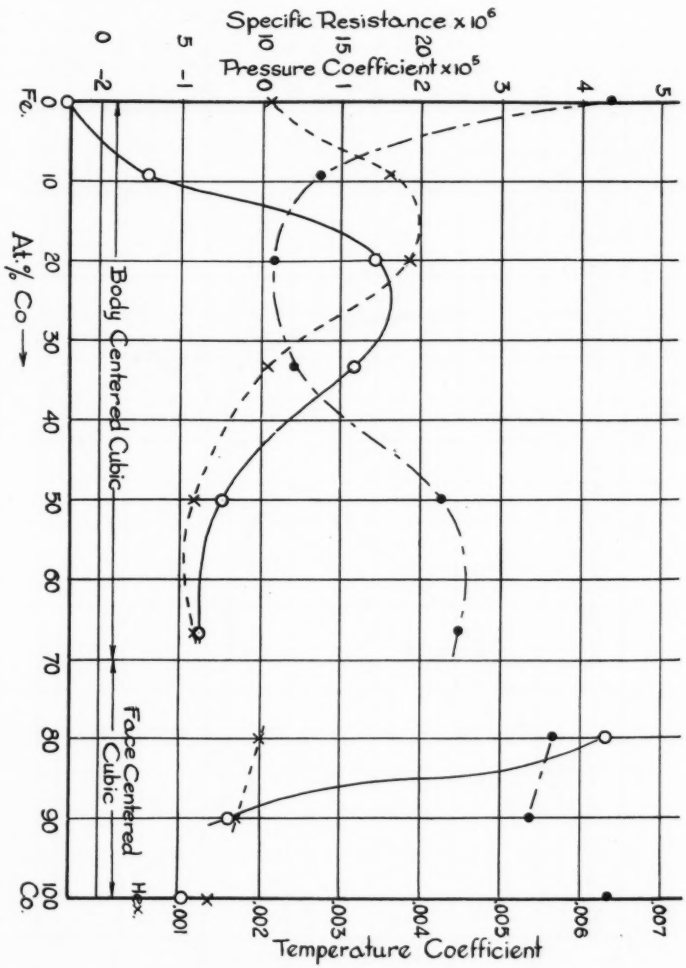

FIGURE 2. Specific resistance at 20° C (dotted line), pressure coefficient of resistance (full line), and temperature coefficient of resistance (dashed and dotted line), in the Fe-Co series as a function of composition.

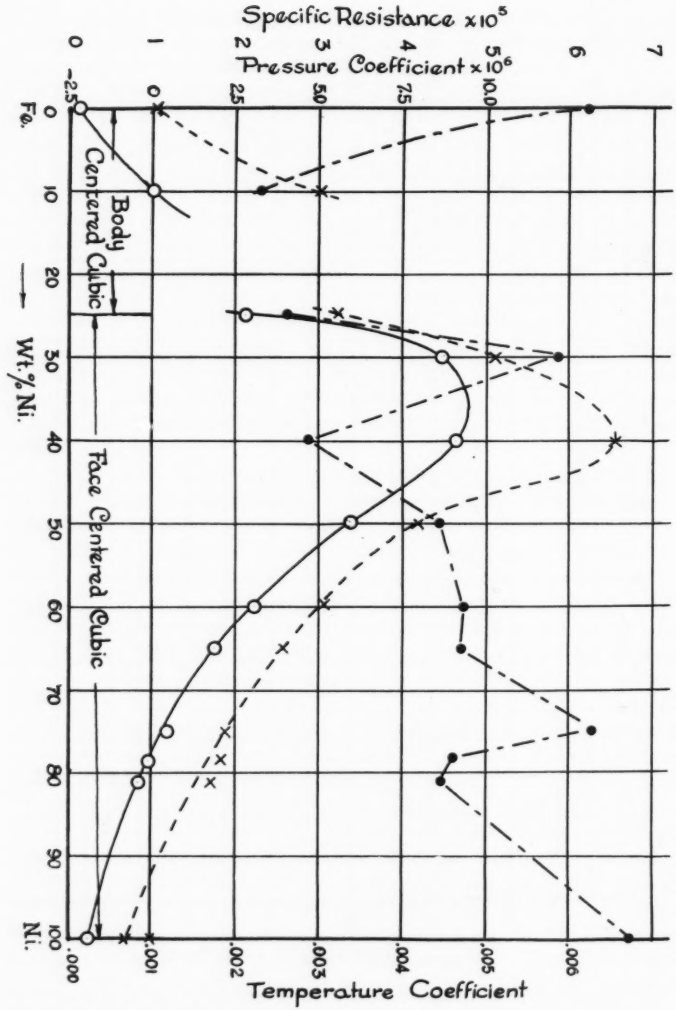

FIGURE 3. Specific resistance at 20° C (dotted line), pressure coefficient of resistance (full line), and temperature coefficient of resistance (dashed and dotted line), in the Fe-Ni series as a function of composition.

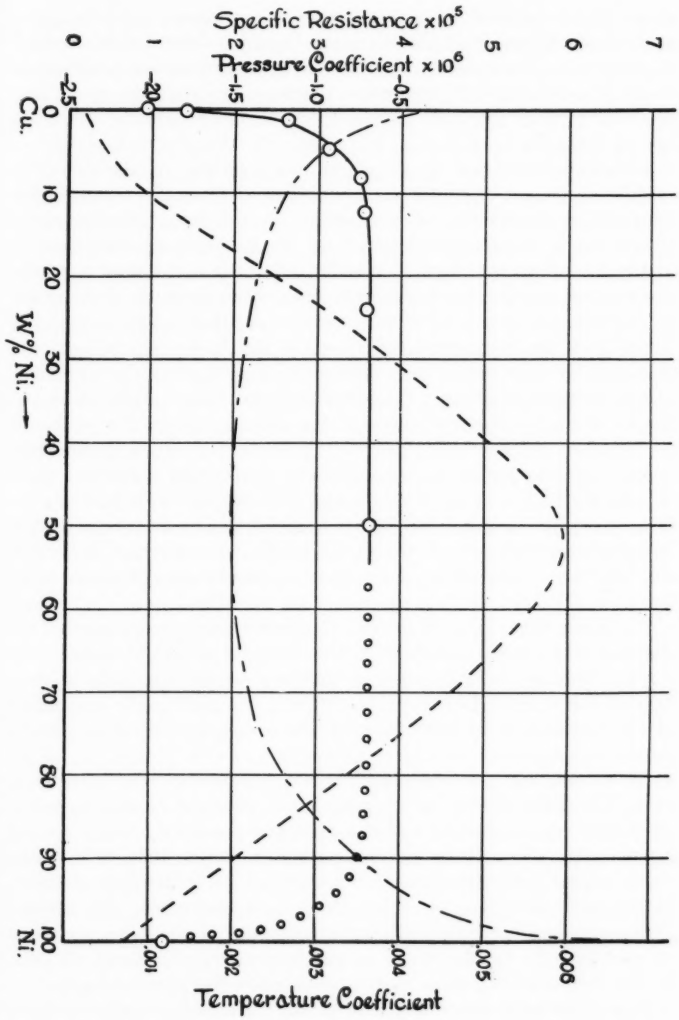

FIGURE 4. Specific resistance at 20° C (dotted line), pressure coefficient of resistance (full line), and temperature coefficient of resistance (dashed and dotted line), in the Cu-Ni series as a function of composition. A hypothetical extension of the curve for pressure coefficient beyond the range of experiment is shown by the small circles.

about by a decrease of external pressure, whereas the change of pressure coefficient is in the direction brought about by an increase of pressure. The same is also true with regard to the addition of Co to Fe, although the numerical discrepancy is not so great, and is probably true, although this is not entirely certain, for the addition of Cu to Ni.

An inspection of the figures shows much greater regularity in the specific resistance and pressure coefficient of resistance than in the temperature coefficient. For example, in the face centered range of the Fe-Ni series, the behavior of the temperature coefficient is highly irregular, but that of specific resistance and pressure coefficient is comparatively regular, both rising from the ends of the range to a maximum at some intermediate composition. This is also the behavior in the body centered range of the Fe-Co series; here the maximum of resistance is more displaced with respect to the maximum of the pressure coefficient than in the Fe-Ni series. The same behavior is shown qualitatively by the Ni-Cu series; the range of variation of specific resistance with composition is here, however, much larger compared to the variation of pressure coefficient than in either the Fe-Ni or Fe-Co series. It would be interesting to know whether the same parallelism holds in other parts of the range, but there are only two points in the body centered range of Fe-Ni, and also only two points in the face centered range of Fe-Co, and two points are not enough to answer the question.

Although there is undoubtedly a parallelism between specific resistance and pressure coefficient, the example of Cu-Ni shows that the parallelism cannot be so close that one can be said to be a function of the other. Between 10 and 50% Ni the specific resistance of Cu-Ni changes six fold, whereas the pressure coefficient changes hardly at all.

An interesting question that cannot be answered at present is as to the discontinuity of resistance and pressure coefficient when the lattice type changes. If the relations previously found for all known cases of melting and polymorphic change hold here,[13] one would expect the resistance to increase, when the lattice type changes, in the same direction in which the volume increases. To answer this, many more alloys must be investigated in the range between 10 and 25% Ni for the Fe-Ni series, and between 66.7 and 80% Co in the Fe-Co series, than were available for the present work.

One of the most interesting results which I hoped to get from these measurements was some suggestion as to the mechanism which is

responsible for a positive pressure coefficient. The conclusion seems to be justified that in these three series of alloys the same thing that tends to cause a high specific resistance also tends to make the pressure coefficient positive. According to the view of conductivity in which I have been interested for some time[14] this would be interpreted in terms of a failure of perfect fit of adjacent atoms. If the atoms do not fit closely together, as they do not in a lattice composed of different sorts of atoms of different sizes, the electrons encounter difficulty in passing from atom to atom and the resistance is high. This is the explanation of the high resistance in the middle of a series of mixed crystals. Now if pressure tends to accentuate the lack of fit, it is to be expected that the pressure coefficient of resistance will be positive. It is natural to expect this to be the case. At pressures of the order of magnitude of those with which we are here dealing, part of the compressibility of a solid comes from the closing of the empty spaces between the atoms, the compressibility of the atoms themselves contributing only partly to the total effect. Hence as pressure increases the relative part of the total volume occupied by the atoms themselves increases, which means that the relative difference between the diameters of the different sorts of atoms is accentuated. But this is merely another way of saying that the lack of fit is accentuated, so that the result to be expected is an increase of resistance, which is the effect actually found.

The general features of this somewhat gross and materialistic point of view can be taken over into the recent picture of conduction on the basis of the wave mechanics given by Houston.[15] According to this picture electrical resistance arises from the scattering by the atoms of the waves which constitute the electrons. If the atoms are spaced with perfect regularity, as in a crystal at 0° Abs., there is little or no scattering, and the resistance is very low. When the arrangement becomes more irregular, as it does when temperature is increased, the scattering increases and resistance increases. If there is a permanent additional cause of irregularity, as when atoms of different sizes are forced to occupy the same lattice in an alloy, there is an additional reason for scattering, which explains the increase of resistance when a foreign metal is added to either pure component. If the irregularity in the space arrangement is accentuated by pressure, as we have just seen is to be expected on geometrical grounds, the scattering is also increased, and the pressure coefficient of resistance tends to become positive.

The reason for the positive pressure coefficient of alloys given in the last paragraph is different from the reason given by Houston for the positive pressure coefficient of pure metals. Houston connects the positive pressure coefficient of pure metals with an abnormally small decrease of compressibility with pressure, or even possibly an abnormal increase of compressibility. It would seem that such a mechanism is not necessary to account for the positive pressure coefficient of alloys. In order to check this point, I have measured by the conventional method, which has now been applied to many substances,[16] the compressibility as a function of pressure of invar, a Fe-Ni alloy of 37.5% Ni content, corresponding approximately to the alloy of maximum resistance and pressure coefficient found above. The following was found for the compressibility at 30° between atmospheric pressure and 12000 kg./cm.^{2}:

$$-\frac{\Delta V}{V^0} = 9.99 \times 10^{-7} p - 6.18 \times 10^{-12} p^2.$$

The second degree term has the normal sign, and numerically is larger, rather than smaller, than that characteristic of pure metals of about the same compressibility. The compressibility itself is unexpectedly large, being 75% greater than the value calculated by the rule of mixtures from the pure components.

It would seem, then, that Houston's explanation of the positive pressure coefficient of pure metals need have no application to alloys. Here we have an adequate picture of the situation in a lack of fit in the crystal lattice, which is accentuated by pressure. The facts disclosed by the compressibility measurement of invar, namely a high compressibility combined with an abnormally rapid decrease of compressibility with pressure, are in complete agreement with this picture.

Houston's explanation of the positive pressure coefficient of pure metals must be taken, I believe, with considerable reservation. His explanation finds a correlation between the positive coefficient and an abnormally small decrease of compressibility with pressure; if examination is made of the behavior of a number of metals, instead of the one or two which Houston considered, it will be found that there is in general no such correlation. It may well be that under the proper conditions we have the same mechanism in pure metals as in alloys, namely a lack of fit accentuated by pressure. This would especially be expected if the atom has not spherical

symmetry. It is perhaps the present tendency, suggested by the wave mechanics, to emphasize the spherical symmetry of the atom, but that the atom cannot always be spherically symmetrical is shown by the mere existence of non-cubic crystals.

I am indebted to my assistant, Mr. Stephen Stark, for making most of the readings in the Fe-Ni and Fe-Co series. I am also much indebted to Mr. Oppenheimer for permission to publish the results which he obtained.

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